

REMARKS

Claims 1-44 are pending in the present application. Claims 2 and 3 have been amended. Support for the amendments can be found in the as-filed specification at least at page 12, lines 17-23; page 22, lines 14-16; and page 29, lines 15-18. No new matter has been added to the application.

Reexamination of the application and reconsideration of the rejections and objections are respectfully requested in view of the above amendments and the following remarks, which follow the order set forth in the Office Action.

Interview

Applicants thank the Examiner for participating in a very helpful and productive interview with Applicants' representative. Applicants include herewith the arguments discussed with regard to the references cited against the currently pending claims.

Rejections under 35 U.S.C. §112

The Office Action maintained the rejection of claims 2 and 3 under 35 U.S.C. §112 for allegedly being indefinite. Applicants respectfully traverse. Applicants have amended claim 2 to recite that the injection system is an automobile heat engine injector. Applicants have amended claim 3 to recite that the injection system is provided with a needle valve. As indicated above, these amendments are fully supported by the as-filed specification. Applicants submit that amended claims 2 and 3 are not indefinite. Accordingly, Applicants request reconsideration and withdrawal of the instant rejection.

Rejections under 35 U.S.C. § 102

First § 102 rejection

The Office Action maintained the rejection of claims 1-5, 8, 10-12, 16-23, 28-31, and 43-44 under 35 U.S.C. § 102(b) as being anticipated by Mayne, et al., *Pyrolytic production of aligned carbon nanotubes from homogenously dispersed benzene-based aerosols*, Chemical Physics Letters; 338: 101-107, ("Mayne"). Applicants respectfully traverse.

Independent claim 1 recites a method for preparing carbon nanotubes or nitrogen-doped carbon nanotubes. The method comprises pyrolysis, in a reaction chamber, of a liquid containing at least one liquid hydrocarbon precursor of carbon or at least one liquid compound precursor of carbon and nitrogen consisting of carbon atoms, nitrogen atoms and

optionally hydrogen atoms and/or atoms of other chemical elements and optionally at least one metal compound precursor of a catalyst metal. The liquid is formed under pressure into finely divided liquid particles by a periodic injection system. The finely divided particles that are formed in this way are conveyed by a carrier gas stream and introduced into the reaction chamber, where the deposition and growth of the carbon nanotubes or nitrogen-doped carbon nanotubes take place.

Independent claim 43 recites a device for carrying out a method for preparing carbon nanotubes or nitrogen-doped carbon nanotubes. The device comprises a reaction chamber in which carbon nanotubes or nitrogen-doped carbon nanotubes are prepared by pyrolysis of a liquid containing at least one liquid hydrocarbon precursor of carbon or at least one liquid compound precursor of carbon and nitrogen consisting of carbon atoms, nitrogen atoms and optionally hydrogen atoms and/or atoms of other chemical elements, and optionally at least one metal compound precursor of a catalyst metal. The device further comprises means for forming said liquid under pressure into finely divided liquid particles, for conveying said finely divided particles by a carrier gas stream and introducing them into the reaction chamber. The means for forming said liquid into finely divided liquid particles, for conveying them and introducing them into the reaction chamber comprise a periodic injection system, provided with an injection head, and a connection ring, in which the carrier gas intake is provided, connecting the injection system to the reaction chamber optionally via an evaporation device.

Mayne discloses a method of producing carbon nanotubes by pyrolyzing homogeneously dispersed aerosols generated from benzene/ferrocene solutions at 800°C or 950°C using a compressed-gas-driven glass atomizer that works on the same principles as a commercial air-driven atomizer. *Mayne*, p. 102, ¶2 and Fig. 1. As can be seen in Fig. 1, the atomizer of Mayne comprised a reservoir containing benzene/ferrocene solution and two orthogonally arranged nozzles (A & B), with nozzle B in fluid communication with the benzene/ferrocene solution in the reservoir. In explaining how the glass atomizer worked to atomize the benzene/ferrocene solutions, Mayne states “[p]assage of a high argon flow ... through nozzle A creates a low pressure before the tip of nozzle B (FIG. 1) thus drawing benzene solution through the orifice (nozzle B). The solution was ‘atomised’ as it emanates ...” P. 102, ¶2. The atomizer worked due to Bernoulli’s principle, i.e., as the velocity increases, pressure falls, thus the high flow rate of argon gas created a low pressure vacuum directly above the tip of nozzle B and effectively pulled or sucked the benzene

solution out of the reservoir in which it was located. In this way, the atomizer of Mayne worked similarly to a conventional automobile carburetor wherein air flows through a venturi shaped pipe at such a velocity that a low pressure vacuum is created in the center of the venturi thereby sucking fuel into the airstream from a nozzle or series of nozzles located in the center of the venturi and positioned orthogonally to the airstream.

In contrast, the periodic injection system of claims 1 and 43 operates similarly to an automobile fuel injection system, wherein fluid is atomized by forcing the fluid through a small nozzle under high pressure. In general, the primary difference between an injection system and a carburetor is that an injection system provides pressure or force from behind the fluid to force the fluid through a nozzle thereby atomizing the fluid and a carburetor creates a vacuum or area of low pressure in front of a nozzle to pull or suck the fluid through the nozzle thereby atomizing the fluid. The theories of operation are fundamentally and completely different.

As the instant specification explains with regard to Fig. 1, the reservoir 1 is subjected to a pressure making it possible to convey the liquid to the injector. *See*, p. 28, ll. 24-31. The pressure may result from the introduction of pressurized gas 5 into the upper part of the reservoir. *See*, p. 29, ll. 1-6. Further, for the injection to be possible, the pressure above the liquid level in the reservoir 1 must be greater than the pressure downstream of the injection system thereby forcing the liquid through the injection system and converting the liquid into a finely divided form. *See*, p. 30, ll. 7-11.

As claims 1 and 43 recite, the liquid is formed under pressure into finely divided liquid particles by a periodic injection system. Mayne fails to disclose or reasonably suggest liquid being formed under pressure into finely divided liquid particles by a periodic injection system. Rather, Mayne discloses liquid being formed into finely divided liquid particles by being pulled or sucked through a nozzle by a pressure vacuum created in front of the nozzle in much the same way as a carburetor.

Applicants further note that claims 1 and 43 recite the use of a periodic injection system. The specification defines "periodic" as generally meaning that the system carries out the injection discontinuously, opens periodically, and preferably operates at a fixed frequency with the opening time and the repetition frequency being adjustable parameters. *See*, p. 12, ll. 8-12. Generally, "periodic" means recurring at intervals of time or occurring or appearing at regular intervals. *See*, <http://dictionary.reference.com/browse/periodic>. All of these definitions require repetition, i.e., more than one cycle. In contrast, Mayne discloses a

continuous atomization of liquid. In particular, Mayne states "after 15 min. at 800°C or 950°C, 'atomisation' was discontinued and the argon flow rate was reduced ..." p. 102, ¶2. While the atomizer of Mayne is turned on and then turned off, a cycle of on and then off without another cycle of on and then off is not periodic. The on/off cycle may be a single period, but it is not *periodic*, as the term is defined in the specification and in general usage.

Accordingly, Applicants submit that claims 1 and 43 are not anticipated by Mayne. Further, because of the fundamental operating differences between the atomizer of Mayne the injection system recited in claims 1 and 43, Applicants submit that one of ordinary skill in the art would have no reason to modify the atomizer of Mayne to change it to a periodic injection system as recited in claims 1 and 43. Therefore, claims 1 and 43 are also not obvious in view of Mayne. Based on the foregoing, Applicants respectfully request reconsideration and withdrawal of the instant rejection.

Second § 102 rejection

Claims 1-5, 8, 10-12, 15-17, 20-23, 28-31, and 43-44 were rejected under 35 U.S.C. § 102(b) as being anticipated by Kamalakaran, et al., *Synthesis of thick and crystalline nanotube arrays by spray pyrolysis*, Applied Physics Letters 2000; 77(21): 3385-3387, ("Kamalakaran"). Applicants respectfully traverse.

Kamalakaran discloses a method of producing carbon nanotubes by spray pyrolysis of ferrocene/benzene solutions in an Ar atmosphere. *Kamalakaran*, p. 3385, ¶2. As seen in Figure 1, the spray pyrolysis device comprised a pyrex nozzle attached at one end to a container for storing and releasing ferrocene/benzene solutions and attached at the other end to a quartz tube. *Id.* at ¶3. The nozzle was contained in a pyrex tube that directed the Ar carrier gas flow around the nozzle. *Id.* The ferrocene/benzene solution was released and atomized into a spray by flowing Ar around the nozzle. *Id.* The spray time lasted between 5 and 15 minutes, depending on the volume of reaction solution and the Ar flow rate. The spray pyrolysis device of Kamalakaran worked in much the same way as the device of Mayne, i.e., a high velocity Ar flow created a low pressure area before the tip of the nozzle thus drawing the ferrocene/benzene solution through the orifice of the nozzle. As explained in detail above for Mayne, with this type of spray pyrolysis device, the liquid is not formed under pressure into finely divided liquid particles by a periodic injection system, as required by claims 1 and 43. Rather, the liquid is drawn through the nozzle by a pressure vacuum before the nozzle. Further, like the Mayne device, the device of Kamalakaran is a continuous

rather than a periodic spray device. As stated in Kamalakaran, the spray lasted between 5 and 15 minutes, depending on the amount of solution and Ar flow rate. Again, as explained in detail above for Mayne, a cycle of on/off is not periodic, as required by claims 1 and 43.

Accordingly, Applicants submit that claims 1 and 43 are not anticipated by Kamalakaran. Further, because of the fundamental operating differences between the spray pyrolysis device of Kamalakaran and the injection system recited in claims 1 and 43, Applicants submit that one of ordinary skill in the art would have no reason to modify the spray pyrolysis system of Kamalakaran to change it to a periodic injection system as recited in claims 1 and 43. Therefore, claims 1 and 43 are also not obvious in view of Kamalakaran. Based on the foregoing, Applicants respectfully request reconsideration and withdrawal of the instant rejection.

Third § 102 rejection

Claims 1-5, 9-12, 15-17, 20-22, 27-31, and 43-44 were rejected under 35 U.S.C. § 102(b) as being anticipated by Terrones, et al., *Novel nanoscale gas containers: encapsulation of N₂ in CN_x nanotubes*, Chem. Commun. 2000: 2335-2336, ("Terrones"). Applicants respectfully traverse.

Terrones discloses a method of producing carbon nanotubes by spray pyrolysis of ferrocene/benzylamine solutions in an Ar atmosphere. *Terrones*, p. 2335. The spray pyrolysis device comprised a long furnace fitted with a quartz tube with one end of the tube being attached to a pyrex cone, which was connected to a container for storing ferrocene/benzylamine. *Id.* at ¶3. A nozzle consisting of a capillary tube was contained in the pyrex cone. *Id.* The solution was released from the container and sprayed into the furnace by flowing Ar around the cone/nozzle area. *Id.* The spraying time lasted between 5 and 10 minutes. *Id.*

The spray pyrolysis device of Terrones works in much the same way as the devices of Kamalakaran and Mayne, i.e., a high velocity Ar flow creates a low pressure area before the tip of the nozzle thus drawing the solution through an orifice. As explained in detail above for Mayne, with this type of spray pyrolysis device, the liquid is not formed under pressure into finely divided liquid particles by a periodic injection system, as required by claims 1 and 43. Rather, the liquid is drawn through a nozzle by a pressure vacuum before the nozzle. Further, like the Mayne and Kamalakaran devices, the device of Terrones is a continuous rather than a periodic spray device. As stated in Terrones, the spray lasted between 5 and 10

minutes. Again, as explained in detail above for Mayne, a cycle of on/off is not periodic, as required by claims 1 and 43.

Accordingly, Applicants submit that claims 1 and 43 are not anticipated by Terrones. Further, because of the fundamental operating differences between the spray pyrolysis device of Terrones and the injection system recited in claims 1 and 43, Applicants submit that one of ordinary skill in the art would have no reason to modify the spray pyrolysis device of Terrones to change it to a periodic injection system as recited in claims 1 and 43. Therefore, claims 1 and 43 are also not obvious in view of Terrones. Based on the foregoing, Applicants respectfully request reconsideration and withdrawal of the instant rejection.

Rejections under 35 U.S.C. § 103

The Office Action sets forth a series of §103 rejections based on the primary references discussed above, Mayne, Kamalakaran, and Terrones, either alone or in combination with additional secondary references or official notice. Applicants respectfully traverse the §103 rejections.

With the exception of the primary reference being different, the grounds of rejection are the same for all of the §103 rejections. As can be seen from the arguments presented above, Applicants arguments with respect to the three primary references are the same, accordingly, in the interest of brevity and reduction of repetition, Applicants will respond to the §103 rejections in masse rather than responding to the rejections based on each primary reference separately. For the sake of clarity, each of the §103 rejections set forth in the Office Action is listed below. Applicants remarks in response to the §103 rejections should be considered to be responsive to all of the §103 rejections listed below.

- Claims 1-8, 10-12, 16-23, and 28-44 were rejected under 35 U.S.C. § 103 as being obvious over Mayne, either alone or in combination with each of the following references, Zhu, et al., Direct Synthesis of Long Single-Walled Carbon Nanotube Strands, Science 2002; 296:284: 884-886 ("Zhu"); Ci et al., Preparation of carbon nanotubules by the floating catalyst method, J. Mater. Sci. Ltrs. 1999; 18: 797-799 ("Ci"); Cassell, et al., Large Scale CVD Synthesis of Single-Walled Carbon Nanotubes, J. Phys. Chem. B 1999; 103: 6484-6492 ("Cassell"); Anderson, U.S. Patent No. 5,697,342 ("Anderson"); Dai WO 00/63115 ("Dai"); and Li et al., Structure and growth of aligned carbon

nanotube films by pyrolysis, Chemical Physics Letters 2000; 316: 349-355 ("Li CPL"); Li et al., Large-Scale Synthesis of Aligned Carbon Nanotubes, Science 1996; 274: 1701-1703 ("Li, Science"); Smiljanic, et al., Growth of carbon nanotubes on Ohmically heated carbon paper, Chemical Physics Letters 2001; 342: 503-509 ("Smiljanic"); Zheng, et al., *Chemical Vapor Deposition Growth of Well-Aligned Carbon Nanotube Patterns on Cubic Mesoporous Silica Films by Soft Lithography*, Chem. Mater. 2001; 13: 2240-2242 ("Zheng"); and official notice with regard to claims 32-42.

- Claims 1-8, 10-12, 16-23, and 28-44 were rejected under 35 U.S.C. § 103 as being obvious over Kamalakaran, either alone or in combination with each of the following references, Zhu, Ci, Cassell, Anderson, Dai, Li CPL, Li Science, Smiljanic, Zheng, and official notice with regard to claims 32-42.
- Claims 1-7, 9-17, 20-44 were rejected under 35 U.S.C. § 103 as being obvious over Terrones, either alone or in combination with each of the following references, Zhu, Ci, Cassell, Anderson, Dai, Li CPL, Li Science, Smiljanic, Zheng, and official notice with regard to claims 32-42.

As indicated in the remarks regarding the §102 rejections, the spray pyrolysis systems disclosed in Mayne (atomizer), Kamalakaran (nozzle sprayer), and Terrones (nozzle sprayer) are completely different in form and functionality from the periodic injection system of the claimed invention. As such, Mayne, Kamalakaran, and Terrones fail to disclose or reasonably suggest a device or a method of using a device wherein liquid is formed under pressure into finely divided liquid particles by a periodic injection system, as required by claims 1 and 43.

With regard to the cited secondary references, Applicants assert that none of the secondary references discloses a periodic injection system wherein the recited liquid is formed under pressure into finely divided liquid particles by a periodic injection system, as recited in claims 1 and 43. As such, none of the secondary references in combination with any of the primary references renders the inventions of claims 1 and 43 obvious. Applicants address each of the secondary references more specifically below.

- Zhu discloses direct synthesis of long strands of single-walled carbon nanotubes by a catalytic chemical vapor deposition technique with a floating catalyst method in a vertical furnace. Zhu fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Ci discloses production of carbon tubules with diameters between 10-100 nm by the floating catalyst method using benzene and hexane as the hydrocarbon liquid. Ci fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Cassell discloses synthesis of single-walled carbon nanotubes by the chemical vapor deposition of methane using optimized catalysts. Cassell fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Anderson discloses a hydraulically-actuated fuel injector with direct control needle valve, especially for a direct-injection diesel-cycle internal combustion engine. C. 3, ll. 3-6. Applicants assert that the technical field of Anderson is far remote from that of the instant invention and thus non-analogous. It should not have been cited against the present application because it is in a field that is different from that of Applicants' endeavor and it would not have commended itself to the attention of one of ordinary skill in the art in considering the claimed invention as a whole. MPEP § 2141.01(a). Further, there is no disclosure in Anderson of other uses or applications for the injector thereof. There is no indication in Anderson that would have led one of ordinary skill in the art to use the injector thereof for other purposes in other technical fields, let alone the very specific technical field of carbon nanotubes and their production. As such, Applicants assert that the conclusion of obviousness based on the combination of Anderson with any of the primary references is based on improper hindsight reasoning, which is forbidden. *See,*

MPEP §2145 (X)(A). The Federal Circuit has cautioned that the determination of obviousness cannot be made by picking and choosing elements from available references using the “blueprint drawn by the inventor” in the application specification. *Interconnect Planning Corp. v. Feil*, 774 F.2d 1132 (Fed. Cir. 1985). Rather, the determination must be made based on the state of the art at the time of the invention. The Court also cautioned “[I]t is impermissible to use the claimed invention as an instruction manual or ‘template’ to piece together the teachings of the prior art so that the claimed invention is rendered obvious. ... This court has previously stated that ‘[o]ne cannot use hindsight reconstruction to pick and choose among isolated disclosures in the prior art to deprecate the claimed invention.’” *In re Fritch*, 972 F.2d 1260 (Fed. Cir. 1992). Because impermissible hindsight reasoning has been used to arrive at this conclusion of obviousness, Applicants submit that the claimed invention is not obvious over any of the primary references in combination with Anderson.

- Dai discloses pyrolyzing hydrocarbon gases in the presence of iron phthalocyanine to produce aligned nanotubes. Abstract and Fig. 1. As discussed in the instant specification, the synthesis technique of Dai involves vaporization of solid iron phthalocyanine stored in a boat placed in a furnace. This method prevents reproducible and stable vapor delivery rates from being obtained. Even in the form of a vapor, it is likewise difficult to convey these products continuously with a constant delivery rate, and this is a problem for application to continuous or semi -continuous production. The claimed method overcomes the problems present in methods such as those disclosed in Dai by using a novel method of carbon nanotube production. Furthermore, Dai fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Li CPL discloses a method for preparation of carbon nanotubes by pyrolysis of iron(II) phthalocyanine, under Ar/H₂ atmosphere at a predetermined temperature using an appropriate substrate in a flow reactor consisting of a

quartz glass tube and a dual furnace fitted with independent temperature controllers. This reference is directed to reporting the detailed growth mechanism of the aligned nanotubes. Li CPL fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.

- Li Science discloses a method for producing ordered, isolated, long carbon nanotubes based on chemical vapor deposition using mesoporous silica containing iron nanoparticles embedded in the pores as the substrate. Li Science fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Smiljanic discloses a method for producing carbon nanotubes using carbon vapor deposition wherein a carbon paper substrate loaded with Ni-Co or Fe catalyst is disposed perpendicular to an outer quartz tube of a reactor. Smiljanic fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Zheng discloses production of carbon nanotubes using carbon vapor deposition. The disclosed method uses a micromolding in capillaries technique combined with three dimensional cubic mesoporous silica films containing iron nanoparticles as catalysts to fabricate carbon nanotubes. Zheng fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Official Notice – The Examiner took official notice that growing nanotubes on a variety of substrates with a catalyst is old and known. Even if such official notice is correct, such official notice still fails to overcome the deficiency in the primary references with regard to the recited periodic injection system.

Thus, the instant claims are not obvious in view of any of the primary references in combination with the Examiner's official notice.

For the foregoing reasons, claims 1-44 are considered allowable. A Notice to this effect is respectfully requested. If any questions remain, the Examiner is invited to contact the undersigned at the number given below.

The Director is hereby authorized to charge any appropriate fees that may be required by this paper, and to credit any overpayment, to Deposit Account No. 23-1925.

Respectfully submitted,

BRINKS HOFER GILSON & LIONE

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By: _____

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